With a process for the direct reduction of particulate iron-containing material by fluidization, reformed gas, at least partially freed from CO₂, is supplied to a fluidized-bed reduction zone as a reducing gas and is carried off from the same as a top gas and at least a portion of the top gas together with reformed gas is utilized for direct reduction. To economize on parts of the plant that are impinged on by reducing gas and in order to achieve savings in terms of heating costs, CH₄ and N₂ are, in addition to CO₂, at least partially removed by adsorption, from 50 to 100% of the reformed gas and 0 to 100% of the top gas, and the tail gas removed from the reformed gas and/or the top gas by adsorption is utilized as a heating gas (Fig.1).
Abstract:

A process for the direct reduction of particulate iron-containing material and a plant for carrying out the process

With a process for the direct reduction of particulate iron-containing material by fluidization, reformed gas, at least partially freed from CO₂, is supplied to a fluidized-bed reduction zone as a reducing gas and is carried off from the same as a top gas and at least a portion of the top gas together with reformed gas is utilized for direct reduction.

To economize on parts of the plant that are impinged on by reducing gas and in order to achieve savings in terms of heating costs, CH₄ and N₂ are, in addition to CO₂, at least partially removed by adsorption, from 50 to 100 % of the reformed gas and 0 to 100 % of the top gas, and the tail gas removed from the reformed gas and/or the top gas by adsorption is utilized as a heating gas (Fig. 1).
A process for the direct reduction of particulate iron-containing material and a plant for carrying out the process

The invention relates to a process for the direct reduction of particulate iron-containing material by fluidization, wherein reformed gas, at least partially freed from CO₂, is supplied to a fluidized-bed reduction zone as a reducing gas and is carried off from the same as a top gas and wherein at least a portion of the top gas together with reformed gas is utilized for direct reduction, and a plant for carrying out the process.

A process of this type is known from EP-A - 0 571 358 and from US-A - 5,082,251. Here, iron-rich fine ore is reduced in a system of serially arranged fluidized bed reactors under elevated pressure by means of a reducing gas formed of natural gas by reforming. The iron powder thus produced is subsequently subjected to hot or cold briquetting.

With these known processes, in order to avoid too high a CO₂ content in the reducing gas, the CO₂ is eliminated from the reducing gas before introducing the latter into the first fluidized bed reactor, through scrubbing by means of a conventional CO₂ scrubbing operation based on the principle of physical or chemical absorption. By the CO₂ scrubbing, a high selectivity is achieved, i.e. predominantly CO₂ and H₂S are removed from the gas stream that is to be purified. Other molecules, such as f.i. H₂, CH₄, N₂ etc., are practically not bound by the absorbent and persist within the purified gas stream.

Natural gas is utilized for heating the reformer and, furthermore, for heating a reducing-gas heater, wherein optionally top gas is admixed.

The invention has as its object to further develop these known processes, namely in that, at constant reducing power, the volume of the reducing gas is to be decreased so as to enable savings in respect of all parts of the plant that are impinged on by the reducing gas. In particular, savings in terms of investment costs for the reducing-gas compressors and a reduction in the electricity consumed in compressing the reducing gas as well as savings with respect to heating costs are to be achieved, while the production capacity is at least to be kept equal or to be increased. Furthermore, it is to be feasible to employ the tail gas forming in the CO₂ elimination plant in a manner that will be useful for the process.

With a process of the initially described kind this object is achieved in that CH₄ and N₂ are, in addition to CO₂, at least partially removed by adsorption, preferably by pressure-swing adsorption, either 50 to 100 % of the reformed gas and 0 to 100 % of the top gas, that the
reducing gas is heated and that the tail gas removed from the reformed gas and/or the top gas by adsorption is utilized as a heating gas.

From DE-C - 40 37 977 it is known per se to alternatively use a CO$_2$-scrubber or a pressure-swing adsorption plant for direct reduction in the direct reduction of lumpy iron ore. With this process, the lumpy iron ore is reduced in a reduction shaft furnace in the fixed bed method and on being yielded from the reduction shaft furnace is charged to a melter-gasifier in which the reducing gas fed to the reduction shaft furnace is produced from coal and oxygen and the reduced iron ore is melted. The process in question is a process that is carried out at low pressure, whereas with the fluidization process in accordance with the invention, the reducing gas is conducted at a pressure that is considerably higher. From this, a particular advantage results for the process according to the invention, namely that - in contrast to the known process - there is no need for additional compressors for conducting the gas to the adsorption process. Accordingly, substantial savings in terms of electric energy result for the fluidization process.

Compared to CO$_2$ scrubbing, the adsorption process opens up the possibility of lowering the specific volume of the reducing gas while maintaining the same level of reducing power, due to the fact that with adsorption considerably larger amounts of CH$_4$ and of inert N$_2$ are sluiced out than compared to CO$_2$ scrubbing. This results in savings with respect to the parts of the plant that are concerned by this circumstance, such as pipes, compressors, valves etc.

Since the tail gas removed from the reformed gas and/or the top gas by pressure-swing adsorption has a very high heating value, it is suitably utilized in order to heat the reducing gas and/or as a heating gas for the reforming process carried out to produce the reformed gas.

Prior to CO$_2$ elimination the reformed gas suitably is cooled down considerably, optionally by mixing with top gas, preferably to a temperature ranging between 20 and 100°C, particularly between 30 and 50°C, whereby it becomes feasible to substantially enhance the efficiency of the pressure-swing adsorption.

The realization of the process according to the invention will be particularly advantageous if the direct reduction of the particulate iron-containing material is carried out in several fluidized-bed reduction zones subsequently connected in series, wherein the fine-particulate iron-containing material is conducted from fluidized-bed zone to fluidized-bed zone by gravity from the top downward and the reducing gas from fluidized-bed zone to fluidized-bed zone in the opposite direction, the reducing gas spent during direct reduction being carried off the
uppermost fluidized-bed reduction zone as a top gas, mixed with reformed gas and utilized as a reducing gas.

Suitably, heating of the reducing gas is effected in two stages, namely in a first stage by heat exchange and in a second stage through partial combustion by means of oxygen that has been introduced the reducing gas. This is of particular advantage if utilizing the pressure-swing adsorption plant, since the pressure-swing adsorption plant yields a reducing gas having 0 Vol.% water. It thus becomes feasible to keep the water content in the reducing gas to a very low level even after heating has been effected by means of afterburning and/or partial combustion, although during afterburning and/or partial combustion the H₂O content increases by 1 to 5 Vol.% at the expense of the H₂ content. Possible losses in CO due to afterburning and/or partial combustion can be balanced by modifying the operating characteristics of the reformer, e.g. using a smaller steam : carbon ratio or f.i. increasing the bypass-stream to the adsorption plant.

A plant for carrying out the process according to the invention, comprising at least one fluidized bed reactor for receiving the iron-oxide-containing material, a reducing-gas feed duct leading to this fluidized bed reactor and a top gas-discharge duct carrying off the top gas forming during reduction from the fluidized bed reactor, with a reformer, a reformed-gas duct departing from the reformer and merging with the top-gas duct, the reducing gas formed from reformed gas and top gas passing into the fluidized bed reactor via the reducing-gas feed duct, and with a CO₂ elimination plant, characterized in that the CO₂ elimination plant is constructed as an adsorption plant, preferably as a pressure-swing adsorption plant, that a duct conducting the gas that has been freed from CO₂ from the adsorption plant to a heating means and a tail-gas discharge duct discharging tail gas that has been separated by the adsorption plant lead to a heating means.

For simple adjustment of the desired chemical composition of the reducing gas, the adsorption plant is bypassed by means of a top-gas branch duct as well as optionally by a branch duct conducting reformed gas, which departs from the reformed-gas duct.

Suitably, the tail-gas discharge duct is flow-connected with the heating means of the reformer or is flow-connected with a gas heater for the reducing gas.

In order to achieve optimum efficiency of the pressure-swing adsorption plant, advantageously a gas cooler is provided in the gas supply duct leading to the adsorption plant.
An optimum design of a plant according to the invention is characterized in that a plurality of fluidized bed reactors is subsequently connected in series, the iron-oxide-containing material being conducted from fluidized bed reactor to fluidized bed reactor via conveying ducts in one direction and the reducing gas from fluidized bed reactor to fluidized bed reactor via connection ducts in the opposite direction, and wherein within each of the fluidized bed reactors cyclones are provided for separating fine particles that have been entrained with the reducing gas.

Efficient heating of the reducing gas is characterized in that there are provided a heat exchanger as a heating means for the reducing gas and in serial arrangement thereto a partial combustion means for the reducing gas with an oxygen-supply duct.

In the following, the invention will be explained in more detail with reference to two exemplary embodiments illustrated in the drawing, Figs. 1 and 2 illustrating one process scheme each, in accordance with a preferred embodiment of the invention.

According to Fig. 1, the plant according to the invention comprises four fluidized bed reactors 1 to 4 consecutively connected in series, wherein iron-oxide-containing material, such as fine ore, through an ore supply duct 5 is supplied to the first fluidized bed reactor 1, in which heating to reduction temperature (or prereduction) takes place, and subsequently is conducted from fluidized bed reactor to fluidized bed reactor via conveying ducts 6. The completely reduced material (sponge iron) is hot briquetted in a briquetting arrangement 7.

If required, the reduced iron is protected from re-oxidation during briquetting by an inert gas system not illustrated.

Prior to introducing the fine ore into the first fluidized bed reactor 1, it is subjected to ore preparation, such as drying and sieving, not illustrated in detail.

Reducing gas is conducted in countercflow to the ore flow from fluidized bed reactor 4 to fluidized bed reactor 3 to 1 and is carried off the last fluidized bed reactor 1, viewed in the gas flow direction, as a top gas through a top-gas discharge duct 8 and is cooled and scrubbed in a wet scrubber 9. Within the fluidized bed reactors 1 to 4, cyclones that are not illustrated in the drawing are provided for separating fine particles entrained with the reducing gas.

The production of reducing gas is effected by reforming in a reformer 10 natural gas fed through a duct 11 and desulfurized in a desulfurization plant 12. The gas leaving the reformer
10 and formed of natural gas and steam essentially consists of H₂, CO, CH₄, H₂O and CO₂. This reformed natural gas is supplied through a reformed-gas duct 13 to several heat exchangers 14, in which it is cooled, water thus being condensed out of the gas.

The reformed-gas duct 13 runs into the top-gas discharge duct 8 after the top gas has been compressed by means of a compressor 15. The mixed gas thus forming is passed through a pressure-swing adsorption plant 16 and is freed from CO₂ and at least partially also from H₂S, CH₄, N₂. It is then available as a reducing gas.

In the adsorption plant 16 including a gas accumulator 16', the gas that is to be purified is conducted through vessels that are filled with molecular sieves; depending on the adsorbent that is selected, certain molecules are removed preferentially, due to their size and polarity. If purifying synthesis gas formed from natural gas by reforming or recycled top gas, selectivity with this process is lower than with CO₂ scrubbing operations, i.e. a more substantial portion of separated gas incurs. However, this waste gas, hereinafter called „tail gas“, in contrast to the waste gas incurring during CO₂ scrubbing has a relatively high heating value and thus can be burned in the furnaces required for the direct reduction process, f.i. for heating the reducing gas or for heating the steam reformer, and can consequently reduce the amount of external energy usually required for combustion.

Via ducts 16", the accumulator 16' for receiving the tail gas is connected with the steam reformer 10 in order to effect the heating of the latter and with a gas heater 19 for heating the reducing gas.

The pressure-swing adsorption plant 16 is arranged to be preceded by a gas cooler 17, in which the gas supplied to the pressure-swing adsorption plant 16 is cooled to approximately 40°C or below, thereby ensuring a good efficiency of the pressure-swing adsorption plant 16. Cooling-down may be effected by direct water cooling or by indirect cooling.

Via a reducing-gas supply duct 18 this reducing gas is heated to a reducing-gas temperature of about 800°C in a gas heater 19 arranged to follow the pressure-swing adsorption plant 16 and is fed to the first fluidized bed reactor 4, viewed in the gas flow direction, where it reacts with the fine ores to produce directly reduced iron. The fluidized bed reactors 4 to 1 are arranged in series; the reducing gas passes from fluidized bed reactor 4 to fluidized bed reactor 3, 2 and 1 through connection ducts 20.
According to the invention, the pressure-swing adsorption plant 16 is either supplied only with reformed gas or with a gas mixture consisting of 50 to 100% of the reformed gas and 0 to 100% of the top gas.

Departing from the reformed-gas duct 13, a branch duct 21 branches off, before the reformed-gas duct 13 merges with the top-gas discharge duct 8. This branch duct 21 unites with the reducing-gas feed duct 18 leading from the pressure-swing adsorption plant 16 to the gas heater 19. Furthermore, a further branch duct 22 departs from the top-gas discharge duct 8 and also unites with the reducing-gas feed duct 18 leading from the pressure-swing adsorption plant 16 to the gas heater 19. By means of these two branch ducts 21, 22, which, obviously, like all other gas ducts, are provided with valves, it becomes feasible either to feed to the pressure-swing adsorption plant 100% reformed gas exclusively or to supply the pressure-swing adsorption plant 16 with a mixed gas consisting of 50 to 100% of the reformed gas and 0 to 100% of the top gas.

In order to adjust a specific analysis of the reducing gas, the following options are available in addition to choosing a specific adsorbent:

- conducting portions ranging from 0 to 100% of the stream of gases conducted to the pressure-swing adsorption plant 16 past the pressure-swing adsorption plant 16 in a defined manner - preferably 0 to 30% of the synthesis gas or reformed gas etc., or preferably 0 to 100% of the recycled top gas.

- due to the fact that in comparison to CO₂ scrubbing the pressure-swing adsorption plant 16 sluices out a greater amount of CH₄ and inert N₂, it becomes feasible to lower the specific volume of the reducing gas while maintaining the same level of reducing power. Thereby it becomes feasible to undertake savings with respect to the parts of the plant that are concerned. If desiring a higher content of CH₄ than can be obtained through the above-described kinds of connections, then via the duct 23 natural gas or pure CH₄ can be introduced into the stream of reducing gas conducted to the reduction reactors 1 to 4.

A portion of the top gas is sluiced out of the gas circulatory system 8 in order to avoid enrichment of inert gases, such as N₂. The sluiced-out top gas is fed through a branch duct to the gas heater 19 for heating the reducing gas and is burnt there. Possible shortages of energy are supplemented by natural gas supplied through a feed duct 24.

The sensible heat of the reformed natural gas emerging from the reformer 10 as well as of the reformer smoke gases is utilized in a recuperator 25 to preheat the natural gas after passage through the desulfurization plant 12, to produce the steam required for reformation and to
preheat the combustion air supplied to the gas heater 19 through duct 26 as well as, if desired, also the reducing gas. The combustion air supplied to the reformer through duct 27 is preheated as well.

In order to avoid a decrease in temperature in the fluidized bed reactor 1 arranged first in direction of the ore flow, it can be of advantage to combust a portion of the reducing gas exiting the second fluidized bed reactor 2 in the first fluidized bed reactor, for which purpose an oxygen supply duct 28 and optionally a natural-gas supply duct 29 open into the first fluidized bed reactor.

In order to keep the reaction temperature in all of the fluidized bed reactors 1 to 4 constant at the same level and thereby achieve a further reduction in the energy demand, hot and fresh reducing gas is supplied to the fluidized bed reactors 1 to 3, which are arranged subsequently to the fluidized bed reactor 4 arranged first in the direction of flow of the reducing gas, directly, via the branch ducts 30, in an amount of approximately 10 % per fluidized bed reactor 1, 2 and 3. Thus, the fluidized bed reactors 1 to 4 are not only connected in series with respect to reducing-gas conduction but, as far as the feeding of a small portion of the reducing gas is concerned, are also connected in parallel, whereas with respect to the discharge or passing-on of reducing gas the fluidized bed reactors 1 to 4 with the depicted exemplary embodiment are exclusively connected in series.

In accordance with the plant illustrated in Fig. 2, heating of the reducing gas is effected in two stages, namely in a first stage by heat exchange in the gas heater 19 and in a second stage through partial combustion in a partial combustion means 31 by means of oxygen that has been introduced into the reducing gas via a duct 32.

Advantageously, the gas that is to be heated is first brought to a temperature ranging from 200 to 600°C in a gas heater 19 that is operated as an indirect heat exchanger. Supply of heat can be effected by burning any desired fuel or fuels, preferably, natural gas and top gas branched off from the reduction process are utilized.

Further heating of the reducing gas to temperatures preferably ranging from 700 to 900°C can be effected by the following variants:
a) by separating a partial stream of the reducing gas and stoichiometrically combusting the same with pure oxygen (optionally it is also feasible to utilize a mixture that contains air). This partial stream is mixed with the remaining, cooler portion of the reducing gas, so that the desired final temperature of the total stream of reducing gas ensues.
b) by introducing the total amount of reducing gas into a combustion chamber and partially
combusting it (i.e. substoichiometrically). By re-mixing the burnt gases with the unburnt
gases, the desired final temperature adjusts.

Thus, by this method of heating, the problem of metal dusting can be eliminated and the
process can be operated in a more economical manner due to the reduced pressure loss
incurring in partial combustion furnaces as compared to typical indirectly heated furnaces.

The analysis of the reducing gas is also modified as a result of the partial combustion; typically,
the content of H₂O increases at the expense of the H₂ content by 1 to 5 Vol.%. The same
applies with regard to the CO₂ content at the expense of the CO content.

In order to adjust a specific analysis of the reducing gas after heating, the process therefore has
to be operated in a suitable manner before heating is effected. This is enabled by variable
operating characteristics of the CO₂ elimination, of the reformer etc.: In this way, CO₂
production during partial combustion can be balanced without difficulty, f.i. by adjusting a
lower CO₂ content at the outlet of the pressure-swing adsorption plant 16. Loss of CO due to
partial combustion can be compensated for by modifying the operational characteristics of the
reformer (f.i. a reduced steam : carbon ratio) or f.i. by feeding a more substantial bypass-
stream to CO conversion. With modifications, the above also applies to the adjustment of the
H₂ and H₂O contents.

Since the pressure-swing adsorption plant produces a reducing gas with 0 Vol.% water, the
water content in the reducing gas passed on to the reduction reactors - i.e. after heating in the
partial combustion furnace - can be kept very low (in the range of 1 to 2 Vol.%).

The invention is not limited to the exemplary embodiments illustrated in the drawing but can be
modified in various respects. For example it is feasible to select the number of fluidized bed
reactors as a function of actual requirements. Instead of pressure-swing adsorption it is also
feasible to employ the temperature-swing adsorption process. The former makes use of the
elevated system pressure of the instant direct reduction process, i.e. adsorption and
regeneration of the adsorbent are effected by different pressurization of the vessels, wherein no
external energy supply is needed for pressurization but the system pressure is exploited
directly. With the second variant, the temperature-swing adsorption (abbr. TSA), the
adsorption- and regeneration process is controlled by a suitable temperature profile at a
virtually constant pressure. The adsorption power of the active medium is a function not only
of the pressure but also of the temperature. With the present field of invention, this process is
regarded as second choice, since pressure-swing adsorption is predestined for the analyses of the reformed gas and the top gas as well as for the present system pressure.

Example A
In a plant corresponding to Fig. 1 and having a capacity per hour of 75 t/h hot briquetted iron, 104 t/h fine ore are reacted.

In the steam reformer 10, 108,000 Nm³/h reformed gas are formed by reaction of 18,100 Nm³/h natural gas with 60,300 Nm³/h steam. The amount of heat required for undergrate firing, namely 94 MW, is covered by natural gas (61 MW), preheated air (23 MW) and tail gas (10 MW).

70% of the reformed gas are mixed with 55% of the recycled topgas and after having been cooled to 40°C are fed to the pressure-swing adsorption plant 16 at a pressure of 14.25 bar. The purified gas, which has a temperature of 45°C and a pressure of 13.45 bar, is mixed with the gas streams conducted past it - in sum 179,900 Nm³/h - and fed to the reducing gas heater 19.

The separated tail gas - 22,900 Nm³/h - is available at a pressure of 0.3 bar, a temperature of 35°C and an energy content of 60 MW.

In order to heat the reducing gas to 835°C, 65 MW are needed, made up of 50 MW tail gas, 14.7 MW preheated air and 0.3 MW top gas.

The hot briquetted iron exhibits a degree of metallization of 92%.

The respective analyses of the gases are:

<table>
<thead>
<tr>
<th></th>
<th>reformed gas</th>
<th>gas to the pressure-swing adsorption plant</th>
<th>tail gas</th>
<th>reducing gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO [Vol.%]</td>
<td>7.7</td>
<td>9.2</td>
<td>9.8</td>
<td>8.8</td>
</tr>
<tr>
<td>CO₂ [Vol.%]</td>
<td>6.0</td>
<td>8.1</td>
<td>38.9</td>
<td>4.1</td>
</tr>
<tr>
<td>H₂ [Vol.%]</td>
<td>47.0</td>
<td>63.1</td>
<td>20.1</td>
<td>67.0</td>
</tr>
<tr>
<td>H₂O [Vol.%]</td>
<td>36.1</td>
<td>1.9</td>
<td>9.8</td>
<td>1.6</td>
</tr>
<tr>
<td>N₂ [Vol.%]</td>
<td>0.9</td>
<td>5.3</td>
<td>4.8</td>
<td>5.7</td>
</tr>
<tr>
<td>CH₄ [Vol.%]</td>
<td>2.3</td>
<td>12.4</td>
<td>16.6</td>
<td>12.8</td>
</tr>
</tbody>
</table>
In comparison to the prior art, the process according to the invention yields a reducing gas that has a markedly lower level of N$_2$ or CH$_4$ (i.e. in accordance with EP-A - 0 571 358: 14.94 Vol.% N$_2$, 16.29 Vol.% CH$_4$) and hence an enhanced reducing power.

The charged fine ore has 96.91 wt.% Fe$_2$O$_3$ and 2.29 wt.% gangue, the balance being L.O.I.

Example B
In a plant corresponding to Fig. 1 and having a capacity per hour of 75 t/h hot briquetted iron, 104 t/h fine ore are reacted.

In the steam reformer 10, 100,100 Nm$^3$/h reformed gas are formed by reaction of 17,200 Nm$^3$/h natural gas with 55,700 Nm$^3$/h steam. The amount of heat required for undergrate firing, namely 86 MW, is covered by natural gas (25 MW), preheated air (21 MW) and tail gas (40 MW).

90% of the reformed gas are mixed with 60% of the recycled top gas and after having been cooled to 40°C are fed to the pressure-swing adsorption plant 16 at a pressure of 14.25 bar. The purified gas having a temperature of 45°C and a pressure of 13.45 bar is mixed with the gas streams conducted past it - in sum 184,000 Nm$^3$/h - and is fed to the reducing gas heater 19.

The separated tail gas - 26,900 Nm$^3$/h - is available at a pressure of 0.3 bar, a temperature of 35°C and an energy content of 87 MW.

In order to heat the reducing gas to 835°C, 68 MW, made up of 47 MW tail gas, 15 MW preheated air and 6 MW top gas, are needed.

The hot briquetted iron exhibits a degree of metallization of 92%.
The respective analyses of the gases are:

<table>
<thead>
<tr>
<th></th>
<th>reformed gas</th>
<th>gas to the pressure-swing adsorption plant</th>
<th>natural gas</th>
<th>tail gas</th>
<th>reducing gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO [Vol.%]</td>
<td>7.7</td>
<td>8.2</td>
<td>0.0</td>
<td>8.8</td>
<td>7.4</td>
</tr>
<tr>
<td>CO₂ [Vol.%]</td>
<td>6.0</td>
<td>6.9</td>
<td>0.2</td>
<td>33.6</td>
<td>2.6</td>
</tr>
<tr>
<td>H₂ [Vol.%]</td>
<td>47.0</td>
<td>60.8</td>
<td>0.0</td>
<td>19.7</td>
<td>64.1</td>
</tr>
<tr>
<td>H₂O [Vol.%]</td>
<td>36.1</td>
<td>1.8</td>
<td>0.0</td>
<td>9.9</td>
<td>1.6</td>
</tr>
<tr>
<td>N₂ [Vol.%]</td>
<td>0.9</td>
<td>4.9</td>
<td>5.3</td>
<td>4.5</td>
<td>5.5</td>
</tr>
<tr>
<td>CH₄ [Vol.%]</td>
<td>2.3</td>
<td>17.4</td>
<td>94.3</td>
<td>23.5</td>
<td>18.8</td>
</tr>
<tr>
<td>C₂H₆ [Vol.%]</td>
<td>0.0</td>
<td>0.0</td>
<td>0.2</td>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

The essential difference between Examples A and B is the different CH₄ content of the reducing gas. CH₄ is regarded as an inert portion for the reduction proper, but still may influence product quality. With Example A, the CH₄ content of the reducing gas is at about 12.8 Vol.%, with Example B, however, at 18.8 Vol.%, which entails a higher carbon content of the subsequent briquetted product. This higher carbon content can be (though not necessarily is) regarded as an advantage with certain applications of the product. For instance, the higher carbon content can lead to energy savings during melting in subsequent steel-making operations carried out in an electric furnace.

The charged fine ore includes 96.91 wt.% Fe₂O₃ and 2.29 wt.% gangue, the balance being made up of L.O.I.

Example C
In a plant for the production of 75 t/h hot briquetted iron the following configuration is selected for producing the reducing gas:
- production of 130,000 Nm³/h reformed gas having the same analysis as in Example A
- 100% of the reformed gas are fed to the pressure-swing adsorption plant 16 after having been cooled
- the gas thus purified is mixed with the recycled top gas
- the sum of the gas streams - 181,000 Nm³/h - is fed to the reducing gas heater 19

The hot briquetted iron exhibits a degree of metallization of 92%.
The respective analyses of the gases are:

<table>
<thead>
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<th>tail gas</th>
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<tbody>
<tr>
<td>CO [Vol.%]</td>
<td>7.7</td>
<td>7.2</td>
<td>7.5</td>
<td>6.5</td>
</tr>
<tr>
<td>CO₂ [Vol.%]</td>
<td>6.0</td>
<td>13.2</td>
<td>61.3</td>
<td>4.4</td>
</tr>
<tr>
<td>H₂ [Vol.%]</td>
<td>47.0</td>
<td>74.4</td>
<td>23.1</td>
<td>70.4</td>
</tr>
<tr>
<td>H₂O [Vol.%]</td>
<td>36.1</td>
<td>0.5</td>
<td>2.5</td>
<td>1.3</td>
</tr>
<tr>
<td>N₂ [Vol.%]</td>
<td>0.9</td>
<td>1.3</td>
<td>1.2</td>
<td>4.2</td>
</tr>
<tr>
<td>CH₄ [Vol.%]</td>
<td>2.3</td>
<td>3.4</td>
<td>4.4</td>
<td>13.2</td>
</tr>
</tbody>
</table>

The iron ore has the same composition as in Example A.

Advantages resulting from this variant:
- reduction in the capacity of the pressure-swing adsorption plant (30 to 40 % as compared to Examples A and B)
- no H₂S content in the tail gas (with Examples A and B, the latter is introduced into the top gas cycle together with the iron ore and in part is sluiced out by the pressure-swing adsorption plant) - as a consequence, SO₂, which is detrimental to the environment, does not incur during subsequent thermal utilization of the tail gas, i.e. a desulfurization plant is no longer necessary.
CLAIMS:

1. A process for direct reduction of particulate iron-containing material by fluidization in which a reformed gas which has been at least partially freed from CO₂ is supplied to a fluidized-bed reduction zone as a reducing gas and is removed from the fluidized bed reduction zone as a top gas and in which at least a portion of the top gas together with the reformed gas is used for the direct reduction, characterized in that:
   - CH₄ and N₂ are, in addition to CO₂, at least partially removed by adsorption from 50% to 100% of the reformed gas and 0% to 100% of the top gas;
   - the reducing gas is heated; and
   - a tail gas removed by adsorption from the reformed gas or the top gas or both is used as a heating gas.

2. The process according to claim 1, characterized in that the adsorption is a pressure-swing adsorption.

3. The process according to claim 2, characterized in that the tail gas is used to heat the reducing gas or to provide heat for a reforming process carried out to produce the reformed gas, or both.

4. The process according to claim 1, 2 or 3, characterized in that prior to CO₂ elimination, the reformed gas is cooled down.

5. The process, according to claim 4, wherein the reformed gas is cooled down to a temperature ranging between 20 and 100°C.

6. The process according to claim 5, characterized in that the temperature is between 30 and 50°C.

7. The process according to claim 5 or 6, characterized in that the reformed gas is mixed with the top gas for cooling down the reformed gas.
8. The process according to any one of claims 1 to 7, characterized in that the direct reduction of the particulate iron-containing material is carried out in several fluidized-bed reduction zones connected in series, wherein the particulate iron-containing material is conducted from one fluidized-bed reduction zone to another by gravity from the top downward and the reducing gas from one fluidized-bed reduction zone to another in an opposite direction, and that the reducing gas spent during the direct reduction is carried off an uppermost of the fluidized-bed reduction zones as the top gas, the top gas being mixed with the reformed gas to form a mixture used as the reducing gas.

9. The process according to any one of claims 1 to 8, characterized in that the heating of the reducing gas is effected first by heat exchange and thereafter by partial combustion.

10. A plant for direct reduction of particulate iron-containing material by fluidization comprising at least one fluidized bed reactor having a gas inlet and a gas outlet, a first gas circulation system connected to the gas inlet and the gas outlet to permit gas to flow through the fluidized bed reactor in a first direction and comprising a gas source, a gas reformer, a carbon dioxide adsorber and a first heater connected in the first gas circulation system, a second gas circulation system comprising the gas outlet, the carbon dioxide adsorber and the gas inlet, and wherein the adsorber includes a discharge duct for a combustible tail gas separated in the adsorber.

11. The plant according to claim 10 further comprising a second heater flow-connected with the discharge duct.

12. The plant according to claim 10 characterized in that the discharge duct is connected to the first heater.

13. The plant according to any one of claims 10 to 12 further comprising a gas cooler in the first gas circulation system and upstream of the adsorber.
14. The plant according to any one of claims 10 to 13 characterized in that the first heater comprises a heat exchanger and a partial combuster.

15. The plant according to any one of claims 10 to 14 characterized in that several of the fluidized bed reactors are connected in series and each of the reactors have inlets and outlets arranged to permit iron oxide containing material to pass through the reactors counter-current to the first direction.